Indian J. Phys. **69A** (2), 237 - 242 (1995)

I J P A
- an international journal

Investigation of acoustical parameters in binary X Li_2O-(100-X)SiO_2 glasses

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Received 8 October 1994, accepted 20 December 1994

Abstract : Ultrasonic velocity and attenuation of binary silicate glass in the system of X Li_2O–(100-X) SiO_2 for various values of X (= 25, 22, 20, 18 and 15) have been performed at a frequency of 5 MHz by the pulse echo-overlap method. The densities of the glasses have also been measured The elastic constant, acoustic impedance, thermal expansion coefficient, packing density, internal friction, Poisson's ratio and microhardness of the glass samples have been calculated from the measured data. The variation of the above parameters with change in composition have been discussed, based on the structural changes of glass network

Keywords : Alkali silicate glass, ultrasonic attenuation, elastic constant, rigidity

PACS Nos. : 43.35.Cg, 61.43.Fs, 62.65.+k

1. Introduction

A growing interest has developed in glasses like borate, silicate, borosilicate and high lead glasses due to its technological applications in various fields [1–5]. The silicate glass system finds application in semiconductor technology, optical communication and other areas of electronics and allied fields [6]. The addition of alkali metal oxides (Li_2O, K_2O *etc*) to silicate glass system eventually changes the glass network, leading to the formation of single-bonded or non-bridging oxygen atoms (NBO). Like other techniques, ultrasonic velocity and attenuation measurements play a significant role in understanding the structural characteristics of glass network. Attempts [7–11] have been made to interpret the variation of ultrasonic velocity and attenuation on glass composition in terms of structural changes of glasses. Further, the derived parameters namely elastic constant, internal friction, thermal expansion coefficient,

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acoustical impedance, packing density, Poisson's ratio, microhardness *etc* as a function of composition of these glasses will give further insight into the rigidity and structure of glasses Recently, a similar type of work in sodium silicate glasses containing another monovalent alkali oxide (K_2O or Li_2O) have been made by Rajendran and El-Batal [12].

It is therefore interesting to measure, the density, velocity and attenuation of some X Li_2O-(100-X)SiO_2 glasses for various values of X (= 25, 22, 20, 18 and 15). Special attention is given to the changes obtained in the various acoustical parameters with the addition of Li_2O in the light of packing state of atom and glass structure.

2. Experimental

2.1 *Glass preparation :*

The different composition of the glass specimens were prepared in an electric furnace using the same technique as reported earlier [13]. The parallelism of the faces of the glass specimens was checked using a micrometer with an accuracy of 2 μm.

2.2. *Measurements :*

(i) *Velocity and attenuation :*

The ultrasonic velocity and attenuation measurements have been carried out simultaneously using ultrasonic time intervalometer (India), based on pulse echo-overlap method [14]. X-cut transducer resonating at a fundamental frequency of 5 MHz was used for the generation and deduction of the longitudinal ultrasonic waves. The transducer was coupled to one end of the faces of the specimens with a good acoustic couplent (Sonotrac G-1) to avoid any air gap between the transmitted pulse and the specimens. The acoustic couplent provide a better impedance matching between the transducer and the specimen as well as reduces the coupling losses. The ultrasonic waves are transmitted through the specimen by the vertical probe 5 MHz Piezo electric transducer. An echo was registered each time, when the transmitted pulses were received by the same transducer after travelling a distance d in the specimen The overlap was achieved for the first two selected echos using the continuous wave (CW) oscillator. The inverse of the frequency gives the travel time (t) in the specimen. Thus, the ultrasonic velocity of the specimen was obtained using

$$U = \frac{2d}{t}. \tag{1}$$

The attenuation coefficient of the specimen was calculated by measuring the amplitude of transmitted (I_0) and received (I) signals of ultrasonic waves as

$$\alpha = \frac{1}{2d} \ln\left[\frac{I_0}{I}\right]. \tag{2}$$

The attenuation coefficient was calculated by repeating the observation one or more time and also by reversing the specimens to interchange the contact faces. The oscilloscope used in this measurement was HCL, India Model 5016.

(ii) Density :

The density of the specimen was measured using Archimedes method in which the specimen was weighed both in air and after immersing in xylene at 293.15 K. The density was calculated using the formula

$$\rho = \frac{a}{(a-b)} \times 0.86 \tag{3}$$

where $\boldsymbol{a}$ and $\boldsymbol{b}$ are the weight of the specimen in air and in xylene and 0.86 is the density of xylene at 293.15 K.

3. Theoretical consideration

The elastic modulus (E) of the glass was related to the longitudinal velocity (U_L) and its density (ρ) as

$$E = U_L^2 \rho. \tag{4}$$

The transmission and reflection of sound energy in the glass specimens was determined using the acoustical impedance

$$Z = U_l \rho. \tag{5}$$

Thermal expansion coefficient can be obtained [14] as

$$\alpha_p = 23.2\,(U_L - 0.57457). \tag{6}$$

The attenuation coefficient and U_L are related to internal friction [15] in a material, for a given frequency by the relation

$$Q^{-1} = \frac{\alpha}{(8.66\,\pi f U_L)}, \tag{7}$$

where α is the attenuation coefficient of the specimen calculated from eq. (2) and f is the frequency of ultrasonic waves.

The packing density [16] is defined by

$$V_T = \frac{\rho}{M} \Sigma V_i x_i, \tag{8}$$

where M is the effective molecular weight, X_i is the mole fraction of the component i and V_i is the packing factor obtained from the following equation for an oxide A_xO_y system,

$$V_i = \frac{6.03 \times 10^{23}\ 4\pi}{3} \left[x R_A^3 + y R_O^3\right] \tag{9}$$

where R_A and R_O are the respective ionic radius of metal and oxygen.

The Poisson's ratio [17] (σ) and microhardness [16] are given by

$$\sigma = 0.5 - \left(1/7.2\ V_T\right), \tag{10}$$

$$H = (1-2\sigma)\,\frac{E}{6(1+\sigma)}. \tag{11}$$

4. Results and discussion

The experimental values of ultrasonic velocity (U_L), density (ρ) and attenuation coefficient (α) of the glass specimens for different compositions are tabulated in Table 1. The

Table 1. Measured longitudinal velocity (U_L), density (ρ) and attenuation coefficient (α) of glasses

Sample No	Composition in weight percent		ρ	U_L	α
	Li_2O	SiO_2	Kgm^{-3}	ms^{-1}	$dB\ m^{-1}$
1	15	85	2305	2487 4	156.7944
2	18	82	2282	2902 6	134 3036
3	20	80	2279	3879 4	98 3214
4	22	78	2270	3807 2	121 7532
5	25	75	2257	3441 7	173 9770

longitudinal elastic modulus (E), acoustic impedance (Z), thermal expansion coefficient (α_p), internal friction (Q^{-1}), packing density (V_T), Poisson's ratio (σ) and microhardness (H) have been calculated and are reported in Table 2.

Table 2. Longitudinal modulus (E), acoustic impedance (Z), thermal expansion (α_p), internal friction (Q^{-1}), packing density (V_T), Poisson's ratio (σ) and microhardness (H) of glasses

Sample No.	$E \times 10^{-9}$ $N\ m^{-2}$	$Z \times 10^{-6}$ $Kg\ m^2\ s^{-1}$	α_p $m\ s^{-1}$	$Q^{-1} \times 10^{10}$ $dB\ s^2\ m^{-2}$	V_T	σ	$H \times 10^9$ Nm^{-2}
1.	14 26	5 7334	57694.3	4 6339	0 5443	0.2448	0 9745
2	19.28	6.6411	67327.0	3 4014	0.5417	0 2436	1 3250
3.	34 30	8.8412	89988.7	1.8631	0.5405	0.2430	2 3639
4.	32.90	8 6423	88313.7	2.3509	0 5393	0.2425	2.2728
5.	26.73	7.7679	79834.1	3.7160	0 5378	0.2417	1 8535

More interesting observation on the addition of Li_2O in X Li_2O–(100-X)SiO_2 glass system has been obtained by analysing the experimental and their derived acoustical parameters. The density shows a continuous decrease (Table 1) with increase in weight percent of Li_2O, whereas the velocity first increases and then shows a maxima at X = 20 and then found to decrease with addition of Li_2O. However, the trend is reversed in the variation of attenuation and it shows a minimum value at the same weight percent X = 20. From the Table 2, it is evident that the effect is well reflected on the derived parameters namely elastic constants, acoustic impedance, thermal expansion coefficient, internal friction and microhardness at X = 20 weight percent of Li_2O.

The initial increase in velocity and decrease in attenuation with addition of Li_2O is caused by the tendency of Li^+ ions to occupy or fill the interstices or empty spaces within the network, each alkali ion is expected to create one non-bridging oxygen (NBO). The creation of NBO in the network is expected to reduce the connectivity. However, Li^+ is known to have stronger bonding to oxygens and contracts the network [18] Also recent structural analysis studies of Li_2O- SiO_2 glasses show [19] phase separation into lithium-rich and silica-rich regions.

Thus, the increase in the values of elastic constants, acoustic impedance, thermal expansion coefficient, is evident from the increase in rigidity of the structure upto X = 20 weight percent of Li_2O. This is further supported by the measured microhardness. On the other hand, the packing density and Poisson's ratio show a continuous decrease with the addition of Li_2O. This results agrees well with the reported results [20–23].

The effect of addition of Li_2O beyond X = 20 weight percent are explained in the following manner. Generally, the addition of alkali oxide to silica causes a splitting of the glassy network composed of Si–O–Si groups and the formation of non-bridging oxygens adjacent to the alkalions [2]. Therefore, with the addition of Li_2O beyond X = 20 weight percent, the formation of non-bridging oxygen (NBO) causes splitting of glassy network. In other words, more alkali ions begin to open up the interstices of *cages* with the network.

Thus, the weakening of the glass structure or a reduction in the rigidity of the glasses beyond X = 20 weight percent results in a decrease in velocity and hence an increase in attenuation was noticed (Table 2). Further, the effect of weakening of glassy structure is well reflected in all other derived acoustical parameters. A similar result have been deduced from the ultrasonic velocity and attenuation measurements in sodium silicate glasses containing alkali metal oxides (Li_2O or K_2O) [12].

The strength of glass network continues to increase upto a least weight percent of Li_2O (X = 20) and in the case of saturation, the non-bridging oxygen atoms (NBO) start appearing, which results in breaking of glass network, Gladkov and Tarasov [24] and Dwivedi *et al* [25] have reported similar anomaly in barium borate and silver borate glasses at about 35% of Na_2O and 25% of Ag_2O respectively.

5. Conclusion

The lithium silicate glasses exhibit a strengthening of the structure of the glass network at lower weight percent (X $\leq$ 20) of Li_2O, while at higher percent (X $\geq$ 20) they exhibit weakening of structure due to the formation of non-bridging oxygen ions. These results are well in turn reflected in the variation of other derived acoustical parameters.

Acknowledgment

One of the authors (V R) is thankful to Prof. G Shanmugam, Principal and The Management, Mepco Schlenk Engineering College for their keen interest in this collaborative work.

References

[1] A Giridhar, S Mahadeven and A K Singh *Bull Mater Sci* **6** 1001 (1984)

[2] K J Singh and S S Bhatti *Acustica* **75** 196 (1991)

[3] M Kodama *J Non Cryst Solids* **127** 65 (1991)

[4] N A Ghoneim, F A Moustafla, A H Zahran and F M Ezz El Din *J Am Ceram. Soc* **66** 447 (1983)

[5] N A Ghoneim, H A El-Batal, Zahran and F M Ezz El Din *Phys Chem Glasses* **24(4)** 83 (1983)

[6] I Fanderlik *Silica Glass and its Applications* (Amsterdam . Elsevier) (1991)

[7] G Carini, Cutionic, M Federico, G Galli, G Tripodo and A Avagadio *Physica* **107B&C** 175 (1981)

[8] S S Bhatti and A P Singh *Acustica* **68** 181 (1989)

[9] Manohar Lal and D P Sing *Indian J Phys* **67** 445 (1993)

[10] A Maiti, S Mukherjee, U S Ghosh and C Basu *J Acoust Soc Indian* **21** 194 (1993)

[11] J Lorosch, M Couzi, J Pelous, R Vacher and A Leasseur *J Non-Cryst Solids* **69** 1 (1984)

[12] V Rajendran and H A El-Batal *Indian J Pure Appl Phys* **33** 29 (1995)

[13] F M Ezz El Din, H A El-Alaity and H A El-Batal *J Radioanlyt Nucl Chem* **163** 267 (1992)

[14] E P Ppadakis *Physical Acoustics* eds W P Mason and R N Thurston (New York Academic) **Vol. 12** (1976)

[15] C A Maynell and G A Saunders *Solid State Commun* **11** 1345 (1972)

[16] D S Sanditov and Sh B Tsdypov *Sov J Glass Phys Chem* **4** 62 (1978)

[17] A Makishima and J D Mackenzie *J Non-Cryst Solids* **17** 147 (1975)

[18] N J Kreidl *Glass Science and Technology* eds D R Uhlmann and N J Kreidl (New York Academic) **Vol. 1** p 107 (1983)

[19] P E Stallworth and P J Bray *Glass Science and Technology* eds D R Uhlmann and N J Kreidl (New York Academic) **Vol. 4B** (1990)

[20] A P Singh, K J Singh and S S Bhatti *J. Pure Appl. Ultrason* **12** 70 (1990)

[21] M A Sidkey, A M Aed-El-Fattah, L Abd-El-Latif and R I Nakhla *J Pure Appl Ultrason* **12** 93 (1990)

[22] J E Shelby *J. Am. Cream Soc* **66** 225 (1983)

[23] A K Varshneya *Fundamentals of Inorganics Glasses* (New York Academic) p 99 (1994)

[24] A V Gladkov and V V Tarasov *The Structure of Glass* (New York Consultants Bureau) **Vol. 11** (1960)

[25] B P Dwivedi, M H Rahman, Y Kumar and B N Khanna *Indian J Phys* **B67** 275 (1993)